Variations in Global Tropospheric OH Over the Last Several Decades

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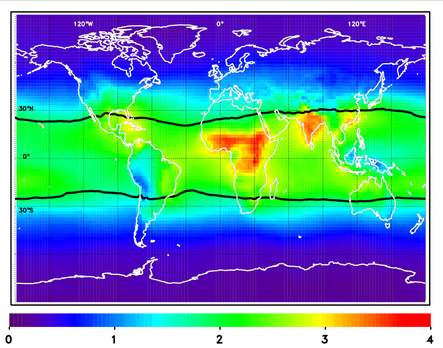
1. Why study OH?

- Hydroxyl radical (OH) is the primary oxidant in Earth's troposphere
- OH is responsible for removing many greenhouse gases (CH_4), ozone-depleting substances (HCFCs & very short-lived halogenated compounds), & pollutants (CO)
- Radical cycling of OH also affects production of greenhouse gas and pollutant tropospheric ozone through reaction with nitrogen oxides (NO_x)

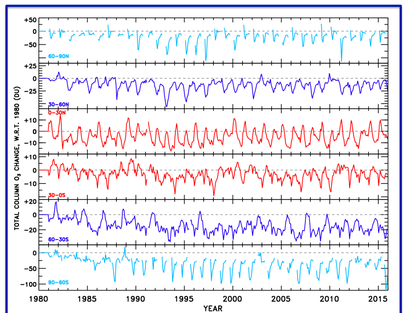


From GMI model, annually averaged for year 1980; black lines mark tropical boundaries

- Short lifetime, high reactivity, & low concentrations of OH make it difficult to measure
- Estimates of global abundances of OH ([OH]^{TROP}), to date, rely on a handful of observable species
- E.g., methyl chloroform (MCF) is lost primarily by OH and is observed by global network
- However, backing out global OH abundances requires assumptions concerning MCF emissions and minor sinks
- Current estimates disagree on how much [OH]^{TROP} varies from year to year
- Attribution of [OH]^{TROP} fluctuations not possible via traditional methods

We aim to answer, using observations of the sources & sinks of OH: How much does [OH]^{TROP} vary over time? What are the drivers of this variability?

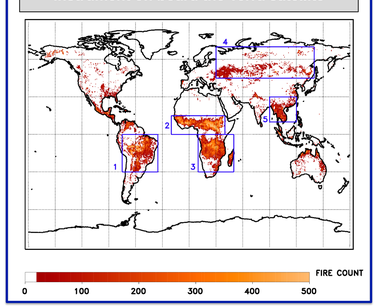
2. Data sets for OH-relevant parameters



Total column O_3 time series from the NASA Solar Backscatter Ultraviolet (SBUV) Merged Ozone Data Set (MOD) data set, utilizing various SBUV, TOMS, and OMI satellite-based instruments

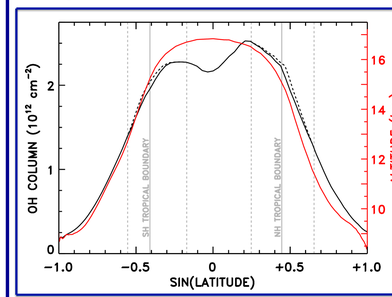
- OH is formed by photolysis of O_3 by UV light followed by reaction with H_2O
- Some other influencers of OH chemistry include temperature, NO_x (secondary production of OH), & CH_4 (loss)
- Sufficient observations exist for most of these factors that we can calculate OH responses to each for years 1980-2015:
  - Overhead O_3: NASA SBUV MOD data set, 1980-2015
  - Water vapor: Atmospheric Infrared Sounder (AIRS) instrument on board NASA Aqua satellite, 2002-2015
  - Temperature: Modern-Era Retrospective analysis for Research and Applications version 2 (MERRA-2), 1980-2015 (also H_2O)
  - CH_4: NOAA Cooperative Global Air Sampling Network, 1983-2015
  - NO_x: Not directly constrained by observations, but fire counts from MODIS on board the NASA Terra satellite show realistic representation of NO_x emissions within the global GMI model

Total fire counts for year 2005 from the MODIS instrument on board the Terra satellite



3. Empirical method for calculating ΔOH

- Initial 3-D field of tropospheric OH concentrations is used from a 1980 simulation of the Goddard Modeling Initiative (GMI) chemical transport model as a starting point (shown as vertically-integrated columns, Box 1)
- Sensitivity of OH to changes in each of the examined parameters (δ(OH)/δ(H_2O), δ(OH)/δ(O^1D) (dependent on overhead O_3), δ(OH)/δ(NO_x), δ(OH)/δ(CH_4), and δ(OH)/δ(T)) is evaluated by box model (DSMACC: *Emmerson and Evans, ACP, 2009*)
- Then, the change in each of the parameters above is found from the observational data set and multiplied by the sensitivity to get a value for Δ[OH]
- For overhead O_3, we use the photolysis model Fast-JX version 7.1 (*Bian & Prather, 2002*) to calculate the change in O_3 photolysis frequency resulting from a change in the overhead O_3 column, since this is the direct mechanism causing OH to vary
- Also included is the impact of Hadley cell expansion (i.e., tropical widening) on OH
- OH concentrations are highest in the tropics due to UV flux and humidity
- Several studies point to expansion of the Hadley cell that is severely underestimated in global climate models (*Allen et al., Nat. Geosci., 2014*)



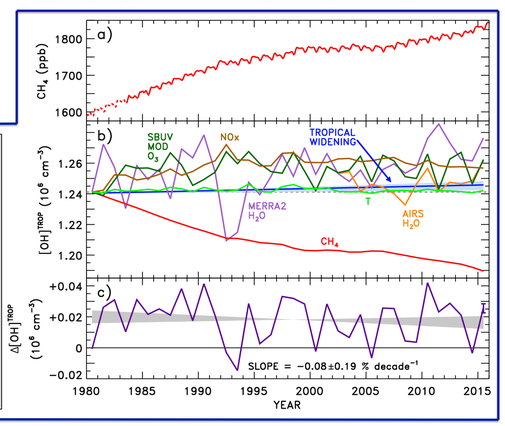
Tropospheric OH Column Distribution across latitudes, shown with (dashed black line) & without (solid black) imposition of tropical widening effect

- If OH increases in the regions that transition from extratropical character to tropical, this impact is missed in model simulations of trends in [OH]^{TROP}
- We include an approximation of this impact by increasing OH in the transition regions to match 1%/decade global rate of widening from the literature

4. Results: What's driving OH variability?

Overall, increases in [OH]^{TROP} due to H_2O, NO_x, overhead O_3, and tropical widening almost balance the decrease expected due to rising CH_4

- CH_4 causes a decrease in [OH]^{TROP} of -1.01 ± 0.05 %/decade, while H_2O has the next largest impact, imparting a $+0.44\pm0.20$ %/decade trend
- Other [OH]^{TROP} trends are: $+0.25\pm0.07$ %/decade due to NO_x, $+0.13\pm0.11$ %/decade due to O_3 column, $+0.12^{+0.07}_{-0.09}$ %/decade due to tropical widening, and -0.02 ± 0.02 %/decade due to temperature
- Total trend in the [OH]^{TROP} anomaly (panel c, below) is -0.08 ± 0.19 %/decade, suggesting *the oxidizing capacity of the troposphere is well-buffered*
- While the effect of tropical widening on [OH]^{TROP} is relatively small, the degree to which global models underestimate this effect is a factor of 4-6, meaning our climate models neglect this contribution to increasing OH

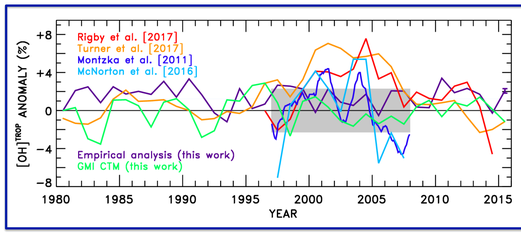


Panel a): evolution of tropospheric mean CH_4 mixing ratio over our study period from NOAA measurements

Panel b): Variations in [OH]^{TROP} calculated in response to time-varying column O_3 (dark green), H_2O (from AIRS post-2002 in orange; from MERRA-2 in purple), NO_x (brown), tropical widening (blue), CH_4 (red), and temperature (bright green)

Panel c): Total change in [OH]^{TROP} calculated as the sum of the effects in panel (b), using only the MERRA-2 source of H_2O data

5. How do these results compare to past studies of OH?



Anomalies (%) in [OH]^{TROP} calculated from this study (dark purple) shown alongside a variety of previous studies and a GMI simulation spanning our study period (green)

- Past studies that perform chemical inversions of MCF and sometimes CH_4 isotopes infer an increase in [OH]^{TROP} in the late 1990s
- Those studies acknowledge assumptions to which their results are sensitive
- Regardless, the pause in CH_4 growth rate has been partly attributed to this increase

Our analysis finds no evidence of this increase in [OH]^{TROP}

- Similarly, a global model simulation of the GMI chemical transport model over this study period shows no discernable trend in [OH]^{TROP}
- We also find differences compared to a study that used chemical transport models to simulate temporally-varying [OH]^{TROP} *(Holmes et al., ACP, 2013)*
- We found a similar impact due to CH_4 as *Holmes et al.*, but a weaker response of [OH]^{TROP} to changes in temperature and H_2O
- While the chemical transport models utilized by *Holmes et al.* may account for feedbacks that we do not capture, our global model simulation suggests similar variability and lack of trend in [OH]^{TROP} to our empirical results

6. Implications

- Understanding how CH_4 emissions are changing is critical for simulating future climate
- While knowledge of anthropogenic emissions is relatively well-constrained, natural emissions from, e.g., wetlands and animal husbandry are difficult to quantify
- Top-down estimates of CH_4 emissions require some knowledge of OH abundances, which may not be sufficiently constrained by a single species observation
- Our study shows that consideration of the major OH sources in addition to its sinks gives different results relative to the most common method of inferring [OH]^{TROP}
- Further work can be done to perform a more complete accounting of [OH]^{TROP} by including CO, volatile organic compounds such as isoprene, and other species that influence OH chemistry; however, constraint of these species to global-scale observations is a challenge

7. Acknowledgments

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- Funding at NASA GSFC was also provided by the NASA Postdoctoral Program, administered by the Universities Space Research Association under contract with NASA
- All data and models used in this work are publicly available through NASA data archives or are otherwise distributed through various web portals
- Further details of this work can be found in the published article, "Changes in Global Tropospheric OH Expected as a Result of Climate Change Over the Last Several Decades," *J. Geophys. Res.–Atmos.*, *123*(18), 2018, doi:10.1029/2018JD028388